

Multi-Triplet Magnons in $\text{SrCu}_2(\text{BO}_3)_2$ Studied by Thermal Conductivity Measurements in Magnetic Fields

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Recently, thermal conductivity in low-dimensional quantum spin systems has attracted interest, because the thermal conductivity due to magnons, κ_{magnons} , has been found to be large in some low-dimensional copper-oxides.¹⁾ Theoretically, the existence of a ballistic κ_{magnon} has been predicted in integrable systems,²⁾ and actually a large thermal conductivity associated with the ballistic κ_{magnon} has been observed in one-dimensional Heisenberg chain systems SrCuO_2 and Sr_2CuO_3 .³⁾ As for non-integrable systems such as spin-gap systems, on the other hand, κ_{magnon} has theoretically been predicted to be diffusive and small. Experimentally, however, a large κ_{magnon} has been observed in a spin-gap state of the two-leg spin-ladder system $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$,⁴⁻⁸⁾ owing to the marked decrease of the magnon-magnon scattering. Therefore, the mechanism of large κ_{magnon} has not yet been understood so clearly.

The compound $\text{SrCu}_2(\text{BO}_3)_2$ is a layered material composed of the two-dimensional CuBO_3 plane, as shown in the inset of Fig. 2(a). The ground state is a spin-singlet state with the spin gap of 34 K⁹⁻¹¹⁾ and can be expressed in terms of the Shastry-Sutherland model.^{9,12,13)} In the spin gap state of $\text{SrCu}_2(\text{BO}_3)_2$, it has been reported that the thermal conductivity due to phonons, κ_{phonon} , is enhanced, owing to the decrease of the phonon-magnon scattering, and that magnons do not contribute to the thermal conductivity.¹⁴⁻¹⁶⁾ This is because single-triplet magnons in the first-excited state are extremely localized, according to the magnetization curve measurements,^{9,10)} the inelastic neutron scattering experiment¹¹⁾ and the perturbation calculations.¹³⁾ On the other hand, the theoretical calculation has suggested that the excited state with higher energy composed of two or several triplet magnons (multi-triplet magnons) can propagate rather fast.^{17,18)} In fact, the existence of such multi-triplet magnons has been confirmed experimentally in the ESR,¹⁹⁾ the inelastic neutron scattering,¹¹⁾ the far-infrared absorption²⁰⁾ and the Raman scattering²¹⁾ measurements. The multi-triplet magnons may contribute to the thermal conductivity when the energy bands

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are lowered by the application of magnetic field. In order to distinguish the contributions of κ_{magnon} and κ_{phonon} from each other, it is effective to study effects of the partial substitution of Zn for Cu on the thermal conductivity, because Zn^{2+} (the spin quantum number $S = 0$) is expected to shorten the mean free path of magnons running in the Cu^{2+} ($S = 1/2$) network.⁷⁾ In this paper, we have measured the thermal conductivity parallel to the a -axis (i.e. parallel to CuBO_3 plane) of the Zn-free and 1% Zn-substituted $\text{SrCu}_{2-x}\text{Zn}_x(\text{BO}_3)_2$ in magnetic fields up to 14 T, in order to examine κ_{magnon} due to the multi-triplet magnons. We have also measured the magnetic susceptibility to check the magnetic properties.

Single crystals of $\text{SrCu}_{2-x}\text{Zn}_x(\text{BO}_3)_2$ with $x = 0$ and 0.02 were grown using the traveling-solvent floating-zone method.^{14,22)} Thermal conductivity measurements were carried out using the conventional steady-state technique. Magnetic fields up to 14 T were applied parallel to the thermal-current direction, using a superconducting magnet. The magnetic susceptibility was measured in a magnetic field of 1 T using a SQUID magnetometer (Quantum Design, MPMS-XL5).

Figure 1 shows the temperature dependence of the magnetic susceptibility, χ , for $\text{SrCu}_{2-x}\text{Zn}_x(\text{BO}_3)_2$. As reported in the previous report,^{9,14)} the susceptibility of $x = 0$ exhibits a peak at 17 K and decreases with decreasing temperature at low temperatures below 17 K, independent of the applied field direction. This is a typical behavior reflecting the formation of spin-singlet dimers with a finite spin-gap. The small Curie-tail observed below 3.5 K may be due to magnetic impurities and/or defects of Cu^{2+} ions. The peak temperature does not change through the 1% Zn substitution, indicating no change in the spin gap. The relative large Curie-tail observed below 6 K may be due to isolated spins which are located adjacent to Zn and able to form no spin-singlet dimers.

Figure 2(a) shows the temperature dependence of the thermal conductivity parallel to the a -axis, κ_a , of $\text{SrCu}_2(\text{BO}_3)_2$ in magnetic fields. In zero field, κ_a exhibits an anomalous enhancement at low temperatures below ~ 10 K, as reported in previous papers.^{14–16,23)} The enhancement in zero field is attributed to the increase of κ_{phonon} , which is caused by the suppression of phonon-magnon scattering owing to the spin-gap formation. The suppression of the enhancement by the application of magnetic field is explained as being due to the reduction of the spin gap. Figure 2(b) shows the temperature dependence of κ_a in the 1% Zn-substituted $\text{SrCu}_{1.98}\text{Zn}_{0.02}(\text{BO}_3)_2$ in magnetic fields. In low magnetic fields below 6 T, it appears that the peak of κ_a in the Zn-substituted sample coincides with that in the Zn-free one. This is because the enhancement of κ_a in the spin-gap state is due to the enhancement of κ_{phonon} and because the spin gap does not change through the 1% Zn-substitution, as mentioned above. In high magnetic fields above 6 T, on the other hand, the peak of the Zn-substituted sample is smaller than that of the Zn-free one. For the clarity, the magnetic field dependence of the relative reduction of the thermal conductivity peak due to the 1% Zn

substitution, $\Delta\kappa_a^{\text{peak}} = [\kappa_a^{\text{peak}}(x = 0.02) - \kappa_a^{\text{peak}}(x = 0)]/\kappa_a^{\text{peak}}(x = 0)$, is plotted in the inset of Fig. 2(b). It is found that $\Delta\kappa_a^{\text{peak}}$ decreases with increasing field at high fields above 6 T while it is negligibly small at low fields below 6 T. This result indicates that multi-triplet magnons contribute to the thermal conductivity above 6 T in the Zn-free sample more than in the Zn-substituted one. In fact, the excitation not only to the first-excited state of single-triplet magnons but also to the higher excited state of multi-triplet magnons has been pointed out to occur from the specific heat measurements.^{24, 25)} That is, the shottky-type peak splits into two peaks around 2–7 K in high fields above ~ 12 T. Therefore, it is concluded that the thermal conductivity peak in high fields above ~ 6 T in the spin-gap state of $\text{SrCu}_2(\text{BO}_3)_2$ is composed of not only κ_{phonon} but also κ_{magnon} due to the multi-triplet magnons.

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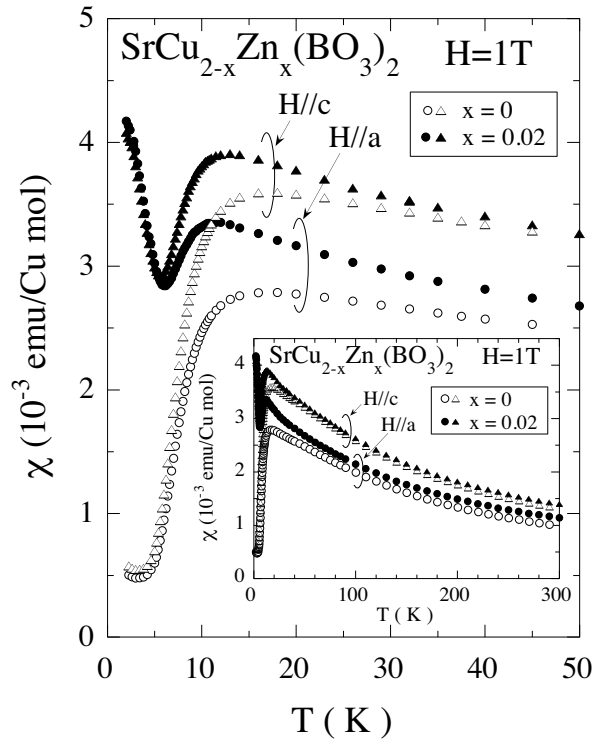


Fig. 1. Temperature dependence of the magnetic susceptibility χ in a magnetic field of 1 T parallel to the a - and c -axes for $\text{SrCu}_{2-x}\text{Zn}_x(\text{BO}_3)_2$ with $x = 0$ and 0.02. The inset shows the data in a wide temperature-region.

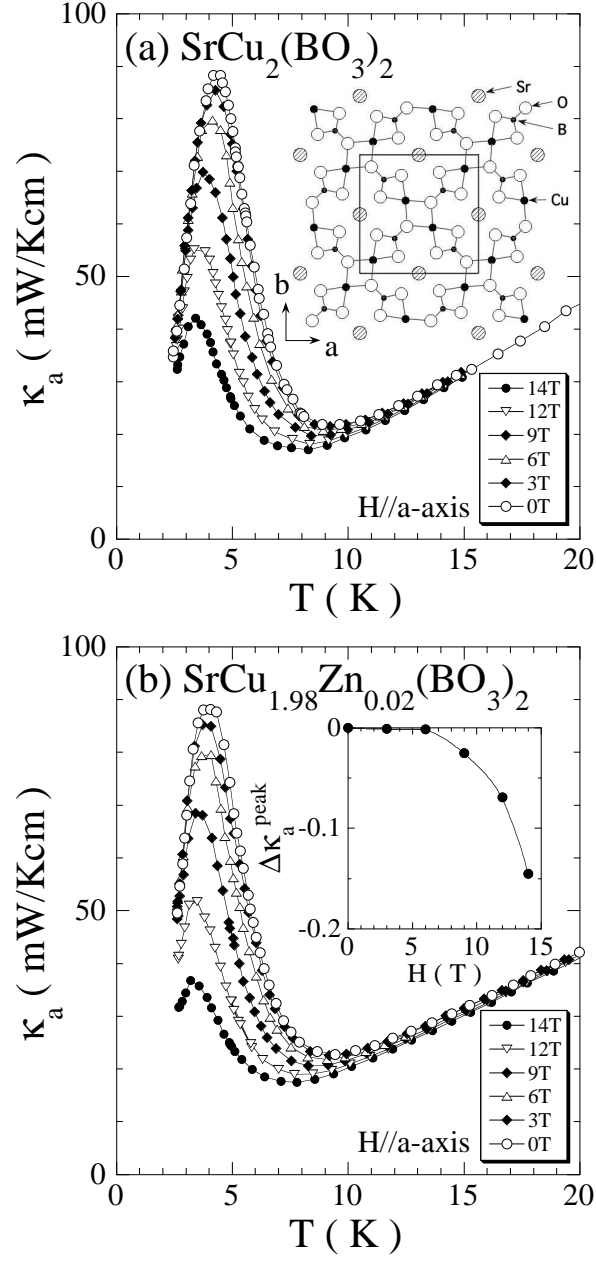


Fig. 2. Temperature dependence of the thermal conductivity parallel to the a -axis, κ_a , of $\text{SrCu}_{2-x}\text{Zn}_x(\text{BO}_3)_2$ with (a) $x = 0$ and (b) $x = 0.02$. The inset of (a) is a schematic picture of the CuBO_3 plane in $\text{SrCu}_2(\text{BO}_3)_2$,⁹⁾ where the solid square indicates the unit cell. The inset of (b) shows the magnetic field dependence of the relative reduction of the thermal conductivity peak due to the 1 % Zn-substitution, $\Delta\kappa_a^{\text{peak}} = [\kappa_a^{\text{peak}}(x = 0.02) - \kappa_a^{\text{peak}}(x = 0)] / \kappa_a^{\text{peak}}(x = 0)$.